



# On the predictions for diffusion-driven evaporation of sessile droplets with interface cooling

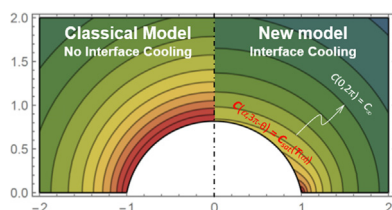
Ha V. Tran, Tuan A.H. Nguyen\*, Simon R. Biggs, Anh V. Nguyen\*

School of Chemical Engineering, The University of Queensland, Queensland 4072, Australia

## HIGHLIGHTS

- Evaporation changes temperature and vapour density at sessile droplet interfaces.
- New predictions for interface vapour density and evaporative flux are obtained.
- New predictions reduce to classical models if interface cooling is negligible.
- Comparison with the literature data shows significant effects of interface cooling.

## GRAPHICAL ABSTRACT



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## ABSTRACT

The diffusion-driven evaporation of sessile droplets from planar surfaces is influenced by cooling at the air-liquid interface. Here, corrections to the available models for predicting the evaporation process are presented. The mass conservation for diffusion-driven evaporation is resolved by considering the effect of interface cooling on the change in density of saturated vapour along the liquid-vapour interface of sessile droplets. Corrections to the predictions for the spatial distribution of vapour density around a sessile droplet and the evaporative flux of vapour at the interface are obtained. The classical models are recovered from the new predictions if interface cooling is negligible. Comparison between the new and classical predictions for the local surface evaporative flux is obtained using the literature data. Our analysis shows a significant effect of interface cooling which should be considered in predicting diffusion-driven evaporation of sessile droplets on planar surfaces.

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## 1. Introduction

Diffusion-driven evaporation of sessile droplets on planar surfaces plays an important role in a number of industrial applications such as the supply of foliar fertilizers, pesticides, and insecticides to plants through the leaf surface, drying of dairy product, spray cooling, ink-jet printing, and coating. Research over the last two decades has focused on understanding how the contact angle and contact line at the intersection between the droplet surface and

the solid surface influence the evaporation kinetics. Droplet evaporation has been explained and described by applying a number of modes (Nguyen and Nguyen, 2012a; Picknett and Bexon, 1977), including the constant-contact-angle, constant-contact-radius, and stick-slip modes.

The underlying theory is based on mass conservation for the vapour evaporation by diffusion, as described by Fick's second law. For example, researchers (Deegan et al., 1997; Erbil, 2015; Picknett and Bexon, 1977) have applied the theory to explain the coffee-ring effect on evaporation of suspensions of colloidal particles. The key equation for a non-uniform local evaporative flux,  $J(\alpha)$ , was obtained as a function of contact angle,  $\theta$ , between the gas-liquid and solid-liquid interfaces as follows:

\* Corresponding authors.

E-mail addresses: [tuan.a.h.nguyen@uq.edu.au](mailto:tuan.a.h.nguyen@uq.edu.au) (T.A.H. Nguyen), [anh.nguyen@eng.uq.edu.au](mailto:anh.nguyen@eng.uq.edu.au) (A.V. Nguyen).

$$J_{\text{classic}} = \frac{C_s - C_\infty}{R/D} \left\{ \frac{\sin \theta}{2} + \sqrt{2}(\cosh \alpha + \cos \theta)^{3/2} \times \int_0^\infty \frac{\tau \cosh \tau \theta}{\cosh \tau \pi} \tanh[\tau(\pi - \theta)] P_{i\tau-1/2}(\cosh \alpha) d\tau \right\} \quad (1)$$

where  $C_s$  is the constant (saturated) vapour density at the droplet surface,  $C_\infty$  is the liquid vapour density far away from the droplet surface (at infinity),  $R$  is the droplet base radius,  $D$  is the vapour diffusion coefficient,  $\tau$  is the integration dummy,  $\alpha$  is the local position of the droplet surface in the toroidal coordinate system, which is related with its radial coordinate (measured perpendicular to the droplet axis of rotational symmetry) by  $r = R \sinh \alpha / (\cosh \alpha + \cos \beta)$ .  $P_{i\tau-1/2}(\cosh \alpha)$  is the toroidal (or ring) function (i.e., the first-kind Legendre function of the complex half-integral degree and the argument of the hyperbolic cosine function) (Magnus et al., 1966) and  $i = \sqrt{-1}$  is the imaginary unit.

Eq. (1) is very well known in the literature and can be found in many papers, e.g., (Hu and Larson, 2002; Nguyen and Nguyen, 2012b; Nguyen et al., 2012; Popov, 2005). It presents one of the key classical models of diffusion-driven evaporation of sessile droplets on a planar surface and has provided the framework for further investigations. Recently, the models have been extended to describe the diffusion-driven evaporation of sessile droplets affected by interface cooling. In this extension, the constant vapour density at the droplet surface, such as  $C_s$  in Eq. (1), is replaced by a function of the local surface coordinate,  $C_s(\alpha)$ , yielding (Gleason and Putnam, 2014)

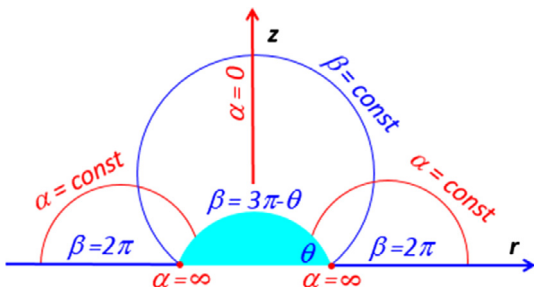
$$J_{\text{CP}} = \frac{C_s(\alpha) - C_\infty}{R/D} \left\{ \frac{\sin \theta}{2} + \sqrt{2}(\cosh \alpha + \cos \theta)^{3/2} \times \int_0^\infty \frac{\tau \cosh \tau \theta}{\cosh \tau \pi} \tanh[\tau(\pi - \theta)] P_{i\tau-1/2}(\cosh \alpha) d\tau \right\} \quad (2)$$

Function,  $C_s(\alpha)$ , of vapour saturation density at the droplet surface in Eq. (2) was determined by interpolating the experimental results for the droplet surface temperature (Gleason and Putnam, 2014).

In this paper, the effect of interface cooling on diffusion-driven evaporation of sessile droplets is re-examined. The new corrections to the available models for diffusion-driven evaporation of sessile droplets are established. We show that the newly established models for describing the diffusion-driven evaporation of sessile droplets affected by interface cooling, such as that described by Eq. (2), have to be corrected.

## 2. Theoretical analysis

We consider a sessile droplet of the spherical-cap shape placed on a solid planar surface. The sessile droplet has rotational symmetry about the direction of gravity. It can suitably be described using



**Fig. 1.** Schematic of the cylindrical co-ordinates  $(r, z, \varphi)$  and the reduced toroidal coordinates  $(\alpha, \beta, \varphi)$  in the meridian plane ( $\varphi = \text{const}$ ) used to describe the evaporation of a sessile droplet with a spherical cap shape on a flat surface. The contact angle  $\theta$  is defined through the liquid phase.

the cylindrical coordinate system  $(r, z, \varphi)$ , whose cylindrical axis ( $z$ ) is opposite to the direction of gravity (Nguyen et al., 2012). The coordinate system has its origin at the centre of the droplet base and its polar axis ( $r$ ) lying on the solid surface (Fig. 1). Mass conservation of vapour evaporation by diffusion is described by Fick's second law. Specifically, the evaporation is usually described by the well-known Laplace partial differential equation,  $\nabla^2 C = 0$ , for the liquid vapour density,  $C$ , in the half-space above the droplet surface and the planar surface. The Laplace equation can suitably be solved by applying the method of separation of variables in the toroidal coordinate system  $(\alpha, \beta, \varphi)$  (Nguyen et al., 2012). The solution can be expressed in terms of the normalized vapour concentration,  $\tilde{C}$ , as follows:

$$\tilde{C} \equiv \frac{C(\alpha, \beta) - C_\infty}{C_e - C_\infty} = \sqrt{2 \cosh \alpha - 2 \cos \beta} \int_0^\infty E_\tau P_{i\tau-1/2}(\cosh \alpha) \cosh[\tau(2\pi - \beta)] d\tau \quad (3)$$

where  $C_e = C_s(\infty)$  is the vapour density at the droplet edge ( $\alpha \rightarrow \infty$ ). In Eq. (3),  $E_\tau$  is a function of the integration dummy (independent of the toroidal coordinates  $\alpha$  and  $\beta$ ), which can be determined from the boundary conditions. The other symbols are previously defined in conjunction with Eq. (1). The solution is independent of  $\varphi$  because of the rotational symmetry. The two coordinate systems are linked by a complex mapping, where  $z + ir = iR \coth \frac{\alpha + i\beta}{2}$ , where  $R$  is the base radius of the droplet. The solution is also bounded by the physical domain:  $\infty > \alpha \geq 0$  and  $3\pi - \theta \geq \beta \geq 2\pi$  as shown in Fig. 1.

Since the complex mapping gives  $r = \frac{R \sinh \alpha}{\cosh \alpha + \cos \beta}$  and  $z = \frac{R \sin \beta}{\cosh \alpha + \cos \beta}$ , we have  $\sqrt{r^2 + z^2} = R \sqrt{\frac{\cosh \alpha + \cos \beta}{\cosh \alpha - \cos \beta}}$ , which shows that a point at infinity (i.e.,  $\sqrt{r^2 + z^2} \rightarrow \infty$ ) is characterised by the "point" of  $\alpha = 0$  and  $\beta = 2\pi$ . The other important details for applying the boundary conditions include the following special values of the toroidal coordinates:

- (1) The droplet edge (i.e.,  $r = R$  and  $z = 0$ ):  $\alpha \rightarrow \infty$  and  $\beta = 2\pi$ ,
- (2) The droplet surface:  $\infty > \alpha \geq 0$  and  $\beta = 3\pi - \theta$ , and
- (3) The solid surface in the vapour phase:  $\infty > \alpha \geq 0$  and  $\beta = 2\pi$ .

It now can be seen that the solution described by Eq. (3) can identically satisfy the following conditions:

- (1) The boundary condition at infinity, i.e.,  $C(0, 2\pi) = C_\infty$  since the term under the square root on the right-hand side of Eq. (3) is equal to 0 when  $\alpha \rightarrow \infty$  and  $\beta = 2\pi$ ,
- (2) The symmetric condition at the axis of symmetry:  $(\partial C / \partial \alpha)_{\alpha=0} = 0$ , and
- (3) The boundary condition at the solid-vapour interface:  $(\partial C / \partial \beta)_{\beta=2\pi} = 0$  for the zero flux of vapour diffusion.

The physical description of the effect of interface cooling during droplet evaporation can be implemented via the boundary condition applied at the droplet surface, i.e.,  $\beta = 3\pi - \theta$ . Traditionally, the vapour concentration at the droplet is considered as a constant as discussed in the Introduction. Due to interface cooling during droplet evaporation, the surface temperature may change along the droplet surface, and so does the surface (saturated) vapour concentration. Therefore, the surface vapour concentration can become a function of the droplet surface coordinates, which is  $\alpha$  in the toroidal coordinate system, and we have

$$C(\alpha, 3\pi - \theta) = C_s(\alpha) \quad (4)$$

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